Remarks

The amendment to the Specification

This amendment is being made to bring the terminology used in Applicants' Specification into better conformity with general technical usage. In general technical usage, a *syntactic foam* is a foam which contains spheres of a material which resists collapse. The purpose of the spheres is to keep the foam from collapsing under pressure. For purposes of the present discussion, syntactic foams can be divided into two classes: *non-expandable* and *expandable*. The syntactic foam described at col. 3, lines 57-59 of Bishop is an example of a non-expandable syntactic foam. As described there, the syntactic foam is "a putty-like mixture of microballoons and adhesive." As Examiner himself agrees, there is no indication whatever in Bishop that Bishop's syntactic foam responds to being heated by expanding beyond the degree of expansion that normally would result from heating the mixture of microballoons and adhesive. (See Examiner's Office action of 10/3/05, par.3)

The syntactic foam used in Applicant's lugs is an example of an expandable syntactic foam. As pointed out at page 7, line 28 of Applicant's Specification, "The particular syntactic foam employed in the preferred embodiment is Loctite SynSpand 9899". A copy of Alex Wong, *Process model for SynsSpand syntactic film*, Technical Paper, Loctite Aerospace accompanies this response. The Introduction to the paper gives an overview of the characteristics and behavior of SynSpand:

> The Loctite SynSpand 9899.1CF is a drapable and pliable epoxy based film which features closed cell expansion. The product expands during heat cure to fill honeycomb core or closed molds. . .

> The patented technology utilizes a closed cell expansion technology which is based upon an expandable polymeric microsphere for homogenous expansion. Expansion of the polymeric microsphere, driven by the encapsulated blowing agent, occurs at approximately 100°C as the shell of the microsphere softens. Expansion is resisted by external pressure and the viscosity of the fluid. As the epoxy resin begins to cure around the expandable polymeric microsphere, it prevents further expansion.

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To clearly distinguish the syntactic foam used in Applicants' lugs from the syntactic foam used in Bishop, Applicants have amended their Specification to replace the term "expanding syntactic foam" with the term "expandable syntactic foam", where "expandable syntactic foam" is intended to mean a syntactic foam that behaves like SynSpand when cured. Because the Specification as filed clearly discloses the behavior of Loctite SynSpand, the amendment adds no new matter. See in this regard page 7, lines 7-30.

The amendments to the claims

The amendment to claim 17

Claim 17 is being amended to point out that the element located between the mold and the tubes is an expandable element, i.e., one that has the function of expanding during the cure, and to point out the effect of the element's expansion on the lay-up. The claim as amended is fully supported by the Specification as filed. See in particular page 6, lines 10-16.

The amendment to claim 18

The amendment is fully supported by the Specification as filed. See p. 6, lines 22-24. As amended, the claim clearly distinguishes between the mold and the silicon which is the expansive element that is located between the mold and the tubes, and thus overcomes the rejection under 35 U.S.C. 112, 2. paragraph.

The amendment to claims 19, 21, and 22

Here, the claim term "expanding foam" has been replaced by "expandable syntactic foam". For the reasons set forth above with regard to the amendment to the Specification, claims 19 and 22 as amended are fully supported by the Specification as filed.

Patentability of the amended claims over Bishop

The disclosure of Bishop

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Bishop discloses a technique for making a bicycle frame out of tubes that are held together by lugs made of woven graphite fiber. The process used to make the frame is described beginning at column 3, line 29. It uses a mold of which one half, indicated by the reference number 20, is shown in FIG. 2. As shown in FIG. 4, to make the frame, you first lay two sheets of impregnated graphite fabric into the hollow provided in the mold for the lug. Then you fill the areas between the hollows with mixture 42, which is a putty-like mixture of microballoons and adhesive. The purpose of adding mixture 42 is to make fillets between the tubes being joined by the lug, as may be seen from col. 2, lines 66-68. The next stage is to put 4 more sheets of fabric into the hollows and over mixture 42 (see FIG. 6). You do the same with the other half of the mold. Then you wrap the frame with two sheets of fabric at the location of the lug. Finally, you put the wrapped frame into the mold, put the other half of the mold on top, and bolt the halves together. The lugs set at room temperature by exothermic action.

Patentability of claim 17 as amended over Bishop

As presently amended, claim 17 reads as follows:

> **17.** (currently amended) A method of making lugs for joints in a bicycle frame made of carbon fiber tubes,
>
> the method comprising the steps of:
>
> making a lay-up of at least carbon fibers and a matrix material around the joint,
>
> applying a mold to the tubes and laid-up fibers and matrix material, and
>
> curing the lug in the mold, the cure including expansion of an expandable element located between the mold and the tubes, the element's expansion serving to compact the lay-up.

The claim now expressly points out that the element that expands during the cure is an *expandable* element, that is, an element that has the function of expanding during the cure, and that the effect of the element's expansion is to compact the lay-up. These limitations serve to clearly distinguish claim 17 from the disclosure of Bishop. In his

rejection of claim 17, Examiner points to the "putty-like mixture 42 of microballoons and adhesive" as the element is located between the mold and the tubes. As pointed out in the discussion of syntactic foam above, mixture 42 of Bishop is a non-expandable syntactic foam and is therefore not an "expandable element", as required by claim 17 as amended. As would be expected by the fact that mixture 42 is not an "expandable element", there is also no indication in Bishop that any expansion that mixture 42 may experience during the cure "serv[es] to compact the lay-up", as further required by claim 17. That being the case, Bishop does not disclose all of the limitations of claim 17 and the claim is consequently patentable over the reference.

Patentablility of the dependent claims

Continuing with the dependent claims, there is no disclosure in Bishop of either the use of a captured silicon mold (claim 18) or the use of a layup including expandable syntactic foam to make lugs (claims 19, 21, and 22), and consequently, these claims are patentable in their own rights over the references. There is further no disclosure in Bishop of the characteristic of the wrapping technique of claim 20 that "the ends of the fabric extend[] beyond the tube" or the characteristic that the ends are wrapped "around the tube the given tube joins to", so claim 20, too, is patentable in its own right over the reference.

Conclusion

Applicant has amended their Specification and claims to conform certain terminology to standard usage and have amended claim 17 to better distinguish it from Bishop and claim 18 to overcome the rejection under 35 U.S.C. 112, second paragraph. The amendments to the claims serve generally to put the claims in better condition for appeal. Moreover, none of the amendments should require a new search. The claims as amended are further completely responsive to Examiner's rejections and are fully supported by the application as filed. Applicant has consequently been fully responsive to Examiner's Office action of 10/03/2005 and Applicant consequently respectfully requests that Examiner enter the amendment and allow the claims as amended.

Respectfully submitted,

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SynSpand®

Technical Paper

PROCESS MODEL FOR SYNSPAND SYNTACTIC FILM

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ABSTRACT

A process model has been developed to simulate the cure behavior of a Loctite patented expandable syntactic film, SynSpand® 9899.1 CF. The model is a PC-DOS based program with graphical results in Windows. It allows the user to input process variables to predict SynSpand performance characteristics. The primary benefit of this model is to provide the user with a tool to minimize the implementation trials and optimize the manufacturing of SynSpand containing parts.

The process model is developed based on fundamental chemical engineering principles and semi-empirical mathematical models on polymer rheology and cure kinetics. SynSpand expansion and the exothermic temperature profile during the cure are predicted by numerically solving the force and energy balances. Chemoviscous effects and cure kinetics of the epoxy are modeled by other semi-empirical constitutive equations.

KEY WORDS: SynSpand, Expansion, Cure Kinetics, Chemoviscosity, Modeling, Expandable Microsphere, Pressure, Heat Transfer

1. INTRODUCTION

The Loctite SynSpand 9899.1CF is a drapable and pliable epoxy based film which features closed cell expansion. The product expands during heat cure to fill honeycomb core or closed molds. The material is used primarily in aerospace structure to fill core to minimize the effects of moisture penetration, reduce weight and provide for localized reinforcement of core. Secondary user benefits include resistance to sonic fatigue, thermal insulation, and structural

reinforcement. Current and future applications range from recreational to aerospace to transportation interior markets.

The patented technology utilizes a closed cell expansion technology which is based upon an expandable polymeric microsphere for homogeneous expansion. Expansion of the polymeric microsphere, driven by the encapsulated blowing agent, occurs at approximately 100°C as the shell of the microsphere softens. Expansion is resisted by external pressure and the viscosity of the fluid. As the epoxy resin begins to cure around the expandable polymeric microsphere, it prevents further expansion.

The SynSpand Process Model has been developed as a predictive tool to shorten the customer's process steps for implementation and optimization of SynSpand applications. With user inputs related to material and processing, the program simulates the SynSpand cure process and predicts the extent of SynSpand expansion and the SynSpand temperature profile during the cure. The model predicted results can then be used as guidelines to predict the cure kinetics, and overfilling or underfilling of SynSpand due to inappropriate cure conditions. The chemical engineering principles used in the model will be discussed in the following sections.

2. THEORETICAL

2.1 BALANCING EQUATIONS

2.1.1 Force Balance

A force balance can be used to describe the expansion of an individual microsphere during the cure of SynSpand. Consider a microsphere being embedded in a fluidized epoxy resin, the force balance can be written in spherical coordinates as:

$$0 = -\delta p/\delta r - [\nabla.\tau]_r \qquad [1]^1$$

where p is the fluid pressure, r is the distance from the center of the sphere, and τ is the stress tensor. From equation of continuity,

$$1/r^2 \, \delta/\delta r \, (\rho r^2 v_r) = 0 \qquad [2]^1$$

where ρ is the epoxy density, and v_r is the velocity of the surrounding epoxy. Upon integration, v_r can be expressed in terms of R_{in}, r, and v_{Rin}, where R_{in} is the inner radius of the microsphere, and v_{Rin} is the velocity of the inner wall of the microsphere. Without considering the inertial term (which is negligible for very small microspheric particles), and assuming there is no interaction between neighboring spheres, the force balance can be simplified to an algebraic equation as follows:

$$P_{int} - P_{ext} - 2\sigma_{in} / R_{in} - 2\sigma_{out} / R_{out} + 4\mu_{wall} R_{in}^2 v_{Rin} (1/R_{out}^3 - 1/R_{in}^3) - 4\mu_{ext} R_{in}^2 v_{Rin} / R_{out}^3 = 0$$
$$[3]^2$$

where P_{int} is the vapor pressure of the blowing agent inside the microsphere, P_{ext} is the applied external pressure, σ_{in} is the microsphere inner wall - blowing agent surface tension, σ_{out} is the microsphere outer wall - epoxy surface tension, R_{out} is the instantaneous outer radius of the microsphere, μ_{wall} is the viscosity of the microsphere wall material, and μ_{ext} is the viscosity of the epoxy. The inertial term can be neglected as long as

$$4\mu_{ext} R_{in}^2 v_{Rin} / R_{out}^3 \gg \rho (3/2 v_{Rin}^2 + R_{in} \delta v_{Rin}/\delta t)$$
$$[4]^2$$

R_{in} at any time t during the expansion can be expressed as a function of R_{out}, assuming that the microsphere remains spherical throughout the expansion and the mass of the polymeric wall material is conserved. R_{out} can be obtained by solving the above force balance. It can then be used to obtain the volumetric expansion of an individual microsphere as a function of time during the cure. The overall expansion of SynSpand is the sum of the expansions of all individual microspheres and can be calculated with known number density of microspheres in SynSpand.

2.1.2 Energy Balance

The temperature profile of the SynSpand part can be predicted by an energy balance written in the following forms:

$$\rho C_{p\text{-}epoxy} dT_{epoxy}/dt = h_{n\text{-}epoxy} (T_n - T_{epoxy}) + \rho (\Delta H_{rxn}) d\alpha/dt$$
$$[5]^3$$
for the SynSpand film

$$\rho C_{p\text{-}i} dT_i/dt = h_{ij} (T_j - T_i)$$
$$[6]^3$$
for other heat transfer layers

The above heat transfer model assumes the SynSpand part is being separated from the oven/heat source by n non-reactive heat transfer layers including air, insulation materials, skin, and tool. T_i and $C_{p\text{-}i}$ denote the temperature and the specific heat capacity of the ith layer, and h_{ij} denotes the heat transfer coefficient between the ith and jth layers. T_{epoxy} and $C_{p\text{-}epoxy}$ denote the temperature and specific heat capacity of the SynSpand layer, and the interfacial heat transfer coefficient between the SynSpand and its adjacent layer is $h_{n\text{-}epoxy}$. Heat transfer coefficients can be determined by monitoring the temperature profiles of the heat transfer layers during an experimental cure run. The heat of reaction ΔH_{rxn} of the epoxy curing can be estimated by DSC. The rate of reaction $d\alpha/dt$ is postulated by the rate equation which will be discussed in the cure kinetics section. The SynSpand temperature profile as a function of time is necessary for calculation of the wall and epoxy viscosities and the vapor pressure of the blowing agent.

2.2 CONSTITUTIVE EQUATIONS

The above balancing equations serve as the backbone for the process model. However, constitutive equations are needed to provide the viscosities, cure kinetics, and vapor pressure of the blowing agent to solve the balancing equations.

2.2.1 Viscosity of the Thermoplastic Microsphere Wall μ_{wall}

The viscosity of the thermoplastic microsphere wall material as a function of temperature can be described by the WLF equation as follows:

$$\log [\mu_{wall}(T) / \mu_{wall}(T_g)] = C_1 (T - T_g) / [C_2 + (T - T_g)] \qquad [7]^4$$

where T_g is the glass transition temperature of the thermoplastic material, and C_1 and C_2 are numeric constants specific to the material. T_g can be determined by Thermomechanical Analysis of raw microspheres. C_1 and C_2 can be obtained by RDS with the thermoplastic wall material. $\mu_{wall}(T_g)$, which is the viscosity of the wall material at its glass transition temperature, is commonly considered to be 10^{13} poise[5].

2.2.2 Chemoviscosity of the epoxy system

The chemoviscosity of a thermosetting epoxy system has been successfully modeled by the Dual Arrhenius equation developed by Martin, Tungare, and Gotro[6].

$$\ln \mu_{ext}(t,T_{epoxy}) = \ln \mu_\infty + (\Delta E_\mu) / RT_{epoxy} + k_x \int^t \exp [- (\Delta E_k) / RT_{epoxy}] dt \qquad [8]$$

where the viscosity of the epoxy is expressed as a function of cure time and epoxy temperature. μ_∞ and ΔE_μ are the preexponential factor and the activation energy for flow respectively. Similarly, k_x and ΔE_k are the preexponential factor and the activation energy for the cross-linking reaction. However, for a more complicated catalyzed system with multiple curatives as in SynSpand, the above equation must be modified to obtain a better model.

The resin system in SynSpand contains an advanced catalytic curing system. At low temperature, reaction is limited to the surface of the catalyst with no extensive cross-linking. As the temperature increases and reaches a critical value, the catalyst begins to melt and initiates the extensive activity of the curing sytem. The basic Dual Arrhenius equation can be modified as follows to obtain a better model for this complicated reaction system.

$$\begin{aligned}
\ln \mu_{ext}(t,T_{epoxy}) = {}& \ln \mu_\infty + (\Delta E_\mu) / RT_{epoxy} + k_{x\text{-}cat} \exp [- (\Delta E_{k\text{-}cat}) / RT_{epoxy}] \\
& + k_{x\text{-}cur1} \int^t \exp [- (\Delta E_{k\text{-}cur1}) / R (T_{epoxy} - T_{cur1})] dt \\
& + k_{x\text{-}cur2} \int^t \exp [- (\Delta E_{k\text{-}cur2}) / R (T_{epoxy} - T_{cur2})] dt \qquad [9]
\end{aligned}$$

The first two terms in the modified equation remains the same as the original equation. The third term describes the localized reaction before the melting of the catalyst. The reaction up to this stage is mass-transfer limited. As the temperature exceeds T_{cur1}, extensive reaction occurs as a result of the activity of the first curative. The chemoviscosity change is described by the fourth

term which is both temperature and time dependent. A similar fifth term is used to describe the chemoviscosity change due to the cross-linking reaction by the second curative which occurs at another temperature of T_{cur2}.

The model parameters can be determined from viscosity data obtained during dynamic cure of the SynSpand base resin system at heating rates of 0.5, 1.0, 2.0, 5.0, and 10.0 °C/min. The predicted and the experimental viscosity profiles for 0.5 and 10.0 °C/min heating rates are shown in Figure 1 below. Notice that a lower minimum viscosity is reached with the higher heating rate because less time is available for the reaction at the higher rate.
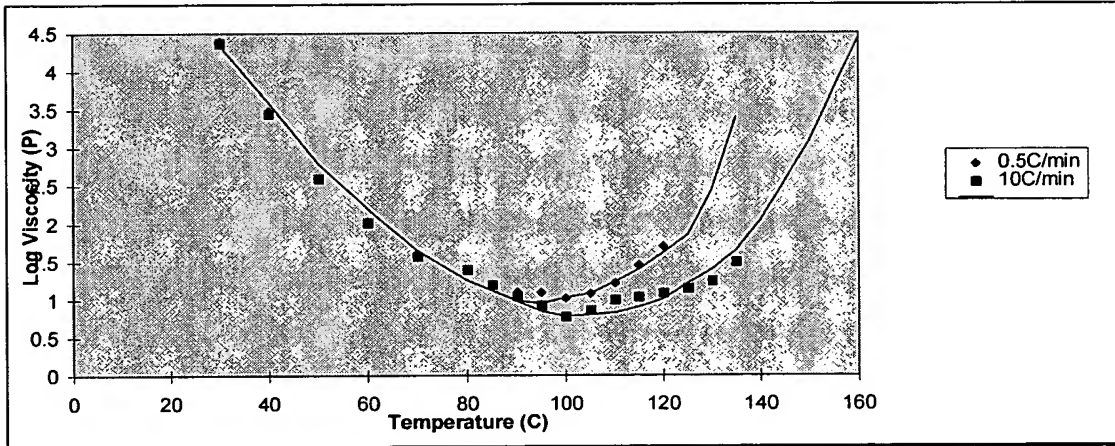
Figure 1. Cure viscosity of the base resin system of SynSpand versus temperature at two different heating rates. Symbols are experimental data by RDS. Solid curves are theoretical predictions by the modified Dual Arrhenius model.

2.2.3 Cure Kinetics of the epoxy system

For epoxy-amine cure reaction, the rate equation has the following general format.

$$d\alpha/dt = (k_i + k_p\alpha)(1 - \alpha)(\gamma - \alpha)$$ [10][7]

where k_p is the catalytic rate constant due to proton donors, k_i is the intrinsic rate constant, and γ is the stoichiometric ratio of amino hydrogens to epoxy groups. This equation is valid for small degree of cure α. For large α however, epoxy curing reaction becomes diffusion limited as the cross-link density increases. The rate equation can be modified as follows.

$$d\alpha/dt = [1/k_p + 1/(k_dD)]^{-1} (\alpha + k_i/k_p)(1 - \alpha)(\gamma - \alpha)$$ [11]

The first term in square bracket in equation [11] is the total resistance toward the completion of the curing reaction. $1/k_p$ is the resistance in the chemical reaction and $1/(k_dD)$ is the resistance in the diffusion of reactive species, where D is the diffusion coefficient as a function of α. This

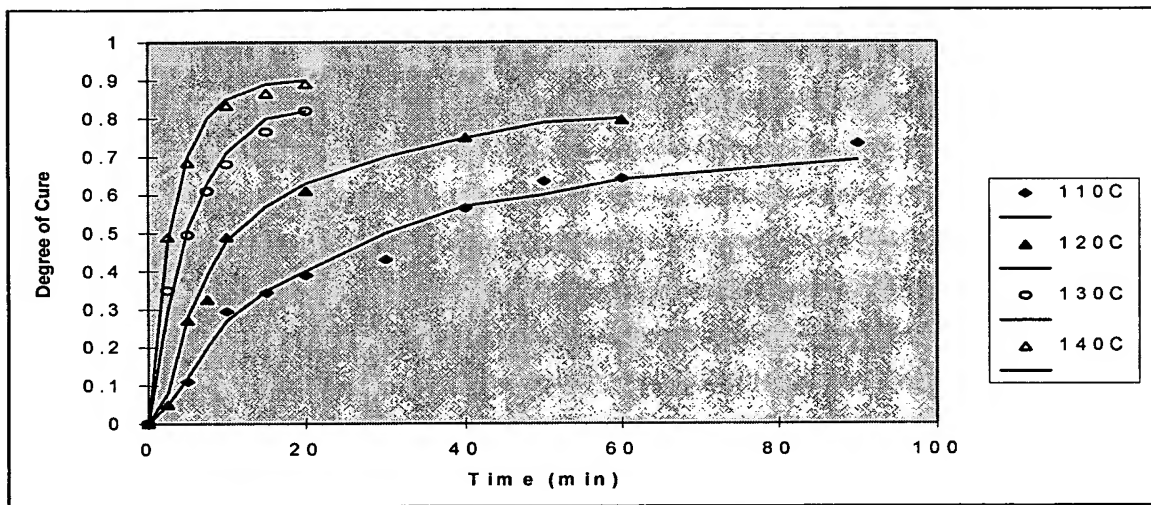diffusion resistance term can be approximated by $A\mu_{ext}$ where A is an empirical parameter. Equation [11] thus becomes:

$$d\alpha/dt = [1/k_p + A\mu_{ext}]^{-1} (\alpha + k_i/k_p) (1 - \alpha) (\gamma - \alpha) \qquad [12]$$

The rate constants can be expressed in an Arrhenius form as follows.

$$k_i = A_i \exp [- E_i / (RT_{epoxy})] \qquad [13]$$
$$k_p = A_p \exp [- E_p / (RT_{epoxy})] \qquad [14]$$

The model parameters and constants can be fitted to isothermal experimental DSC data covering from 110°C to 140°C. Figure 2 below shows the comparison between theoretical predictions and experimental results.

3. EXPERIMENTAL

3.1 THERMAL MEASUREMENT

Thermal properties of the SynSpand product and its base resin system during the cure was measured by a Perkin-Elmer Differential Scanning Calorimeter (DSC) model DSC-7. A few milligrams of samples were weighed onto a small aluminum panel and dry nitrogen was flowing continuously over the sample. The heat of reaction ΔH and the glass transition temperature T_g of the sample can be determined by either isothermal or dynamic curing conditions.

The glass transition temperature of the microsphere wall material was determined by thermomechanical analysis (TMA). A quartz expansion probe was used with a small amount of microspheres spread over the bottom of a stainless steel pan producing a sample thickness of

approximately 0.14mm. Samples were heated at various heating rates in a nitrogen atmosphere. T_g of the thermoplastic wall material was determined as the onset temperature of expansion.

3.2 CHEMOVISCOSITY MEASUREMENT

A Rheometrics System RDS 2 was used to measure the chemoviscosity of the base resin system of SynSpand. The sample was held between two parallel plates enclosed in a heat chamber. The plates were set in oscillation at a frequency of 10 rad/s. The force required to sustain the oscillation was measured by a torque transducer and the results can be translated to complex viscosities.

4. NUMERICAL METHOD

The Runga-Kutta method was employed to solve the equations simultaneously[9]. The program steps through time and numerically integrates the equations to obtain the instantaneous expansion and SynSpand temperature at any time during the cure. Since most changes occur within a narrow window in the cure cycle starting from the glass transition temperature of the microsphere wall material to the onset of the extensive cross-linking of the epoxy system, a variable time-step technique was employed. At the beginning of the cure when little changes occurred, a large time-step was used. Once expansion was initiated when abrupt changes occurred, the time-step was adjusted automatically according to the rate of expansion. When the reaction became diffusion controlled and extensive cross-linking was formed, the time-step was increased back to a large value.

The program code was written in FORTRAN. It consisted of a hierarchy of modular subroutines and functions to facilitate future modifications and extensions.

Figure 2. Degree of cure of the base resin system of SynSpand versus cure time at four different temperatures. Symbols are experimental data by DSC. Solid curves are theoretical predictions based on [12].

2.2.4 Vapor Pressure of the Blowing Agent inside the Polymeric Microsphere

The vapor pressure of the blowing agent is the main driving force for the expansion of SynSpand. It is represented by the Antoine equation below[8].

$$\log P_{int} = A - B / (T + C) \tag{15}$$

The constants A, B, and C are specific to the blowing agent and can be obtained from standard reference.

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The program code was written in FORTRAN. It consisted of a hierarchy of modular subroutines and functions to facilitate future modifications and extensions.

5. MODEL PREDICTED RESULTS AND APPLICATIONS

The model requires the user to input cure parameters related to heating rates, cavity or core depth for the SynSpand expansion, the quantity of SynSpand, and heat transfer conditions such as specific heat capacity and mass of tool, thickness of insulation materials, and so on. The model will then generate output results of the SynSpand part temperature during the cure and the extent of SynSpand expansion. The output results can be used to predict whether an underfilling, overfilling, or complete fill of SynSpand has occurred, and/or the SynSpand has reached a temperature too high that risks a potential runaway reaction. The user can then re-run the model with adjusted cure parameters based on these information. Figure 3 is a flow chart that shows the input and output parameters of the model. Figure 4 is a guide that shows the user how to adjust the input parameters based on model predicted results. Figures 5 to 8 show the output results of two simulated cure cycles. The model predicts a complete fill for the first cycle and an underfill for the second cycle.

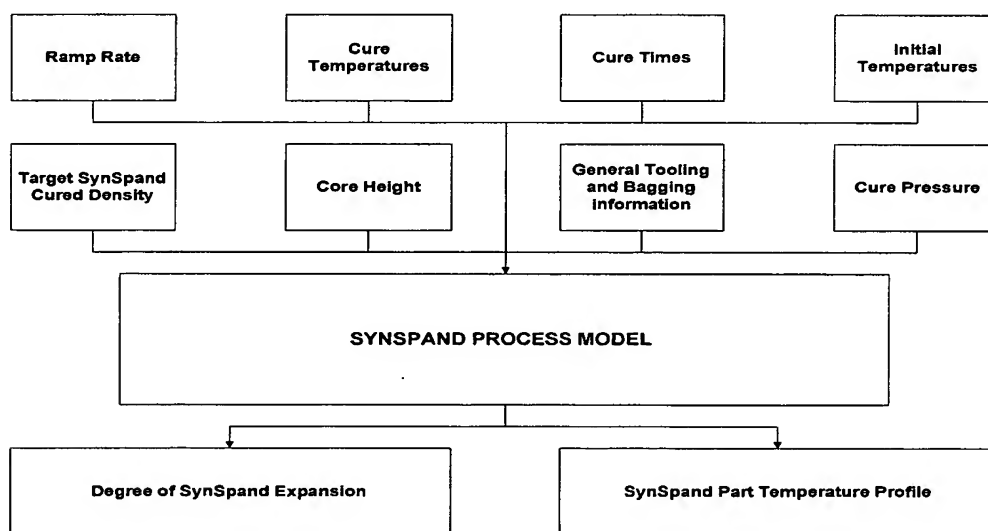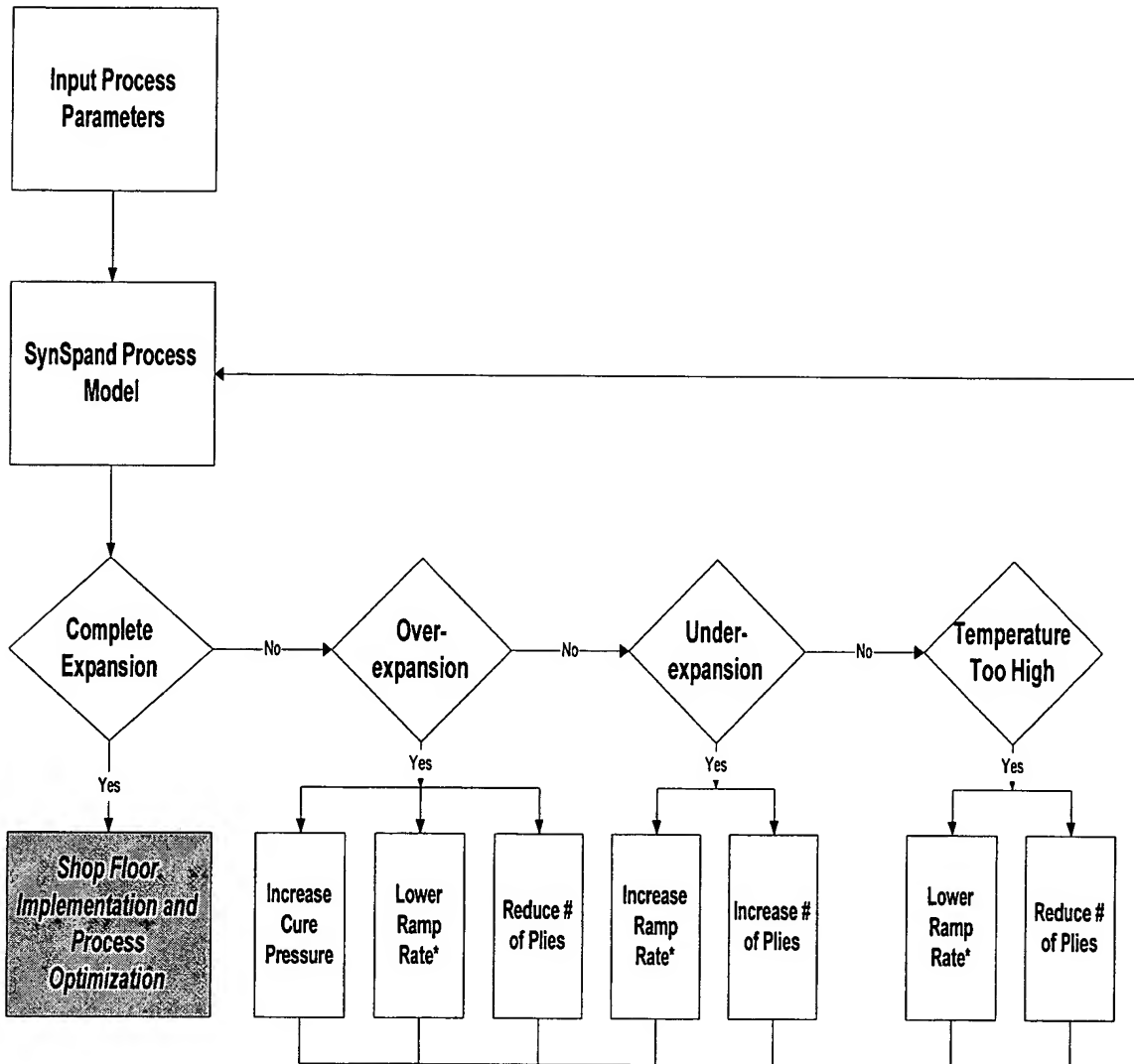**Figure 3. Input and Output Variables of SynSpand Process Model**

Figure 4. A Quick Guide to the SynSpand Process Model

* Effective ramp rate can be adjusted by changing the tooling and bagging inputs

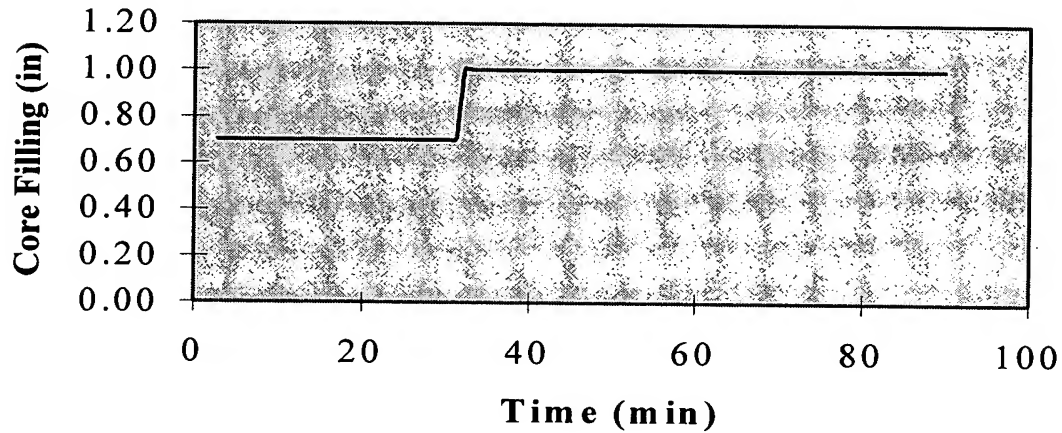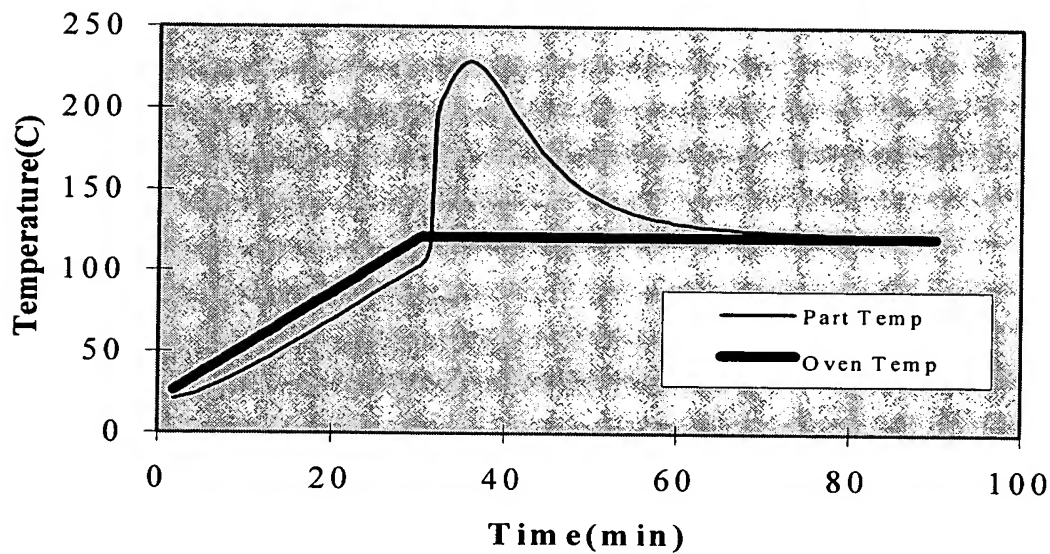## Figure 5. Core Filling (in)

Figure 6. Part and Oven Temperatures

Figures 5 and 6. Simulation results of a SynSpand cure in an autoclave at 100 psi, target cured density of 29 lb/ft³, ramp rate of 6°F/min to 250°F, and expansion into a 1 in cavity. Figure 5 shows the corefilling by the SynSpand and figure 6 shows the SynSpand temperature profile. Notice that the core is completely filled up to 1 in. at the end of the cure.
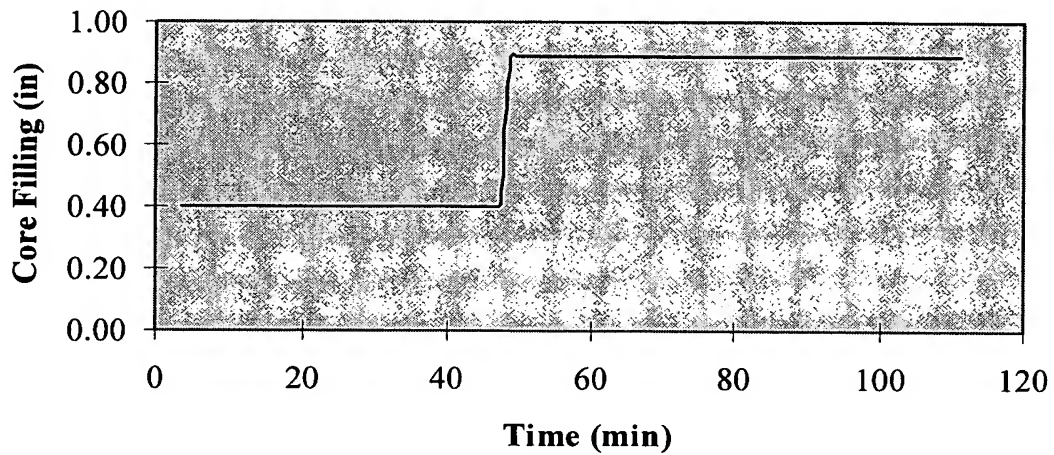
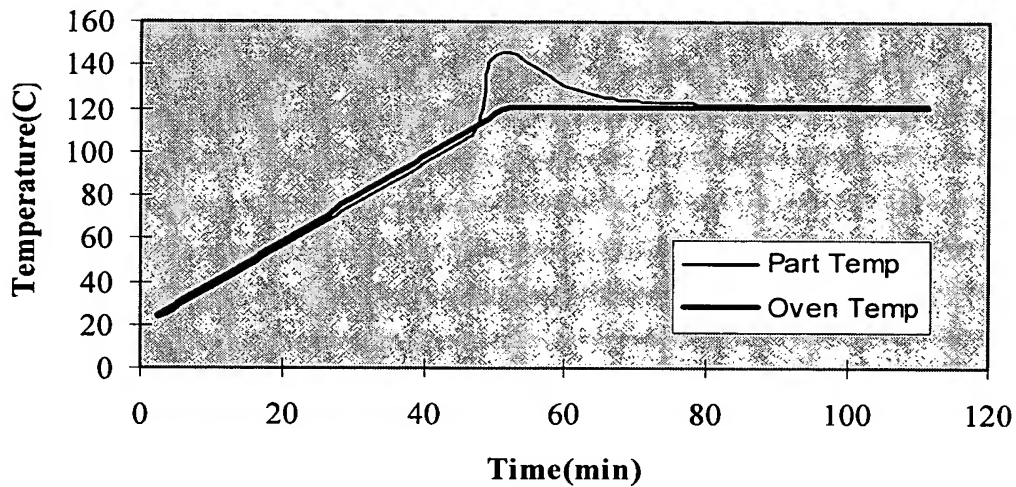## Figure 7. Core Filling (in)

Figure 8. Part and Oven Temperatures

Figures 7 and 8. Simulation results of a SynSpand cure in an autoclave at 100 psi, target cured density of 18 lb/ft³, ramp rate of 3.5°F/min to 250°F, and expansion into a 1 in cavity. Figure 7 shows the corefilling by the SynSpand and figure 8 shows the SynSpand temperature profile. Notice that the core is not completely filled and the peak temperature is substantially lower for a low density cure at a lower ramp rate.

The model predicted results agree well with the experimental data. The following table shows the comparison between the experimental obtained peak exothermic temperature of the SynSpand part and the percent of core filling with the predicted values. The experiment was performed in a closed mold with standard nomex core with 3/8" cells at a typical ramp rate of 6°F/min. The variables are the mold height and the initial thickness of SynSpand.

Initial SynSpand Thickness (in)	Mold Height (in)	Experimental Peak Exotherm (°C)	Theoretical Peak Exotherm (°C)	Experimental % Core Filling	Theoretical % Core Filling
0.2	0.5	134	141	100	100
0.4	0.5	142	149	100	100
0.4	1.0	176	152	100	100
0.8	1.0	231	237	100	100
0.5	1.25	209	209	92	99
0.8	1.25	223	237	100	100
1.0	1.25	244	252	100	100
0.8	2.0	258	237	80	90

6. CONCLUSION

A model has been developed to simulate the curing process of a Loctite expandable syntactic film product SynSpand 9899.1CF. The model is developed based on fundamental force and energy balancing equations. Constitutive equations such as WLF equation, modified Dual Arrhenius model, and diffusion limited cure kinetics model were also used. The model can be used to predict the expansion and temperature profile of SynSpand during the cure at various process conditions. Model results can facilitate SynSpand users in the implementation of the product by reducing the number of shop floor trials.

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